

# PATENT ABSTRACTS OF JAPAN

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(71)Applicant : MATSUSHITA ELECTRIC IND CO LTD

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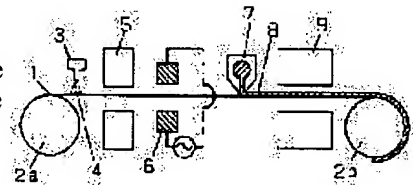
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## (54) MANUFACTURE OF ELECTRODE FOR BATTERY AND BATTERY HAVING ELECTRODE MADE BY THIS METHOD

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To sharply restrain the degradation of charging-discharging capacity of a battery using this electrode and the degradation of a load characteristic by spraying titania and a silicon inorganic material on the surface of a collector, and performing light irradiating processing after it is dried before slurry containing an electrode active material is applied to the collector.

**SOLUTION:** A mixture 4 of titania and a silicon inorganic material is sprayed on a surface of metallic foil 1 supplied from reel 2a. The sprayed mixture 4 is dried by a drier 5. Next, light irradiating processing is performed by corona discharge or the like by using a light radiator 6. By using a coater 7, slurry 8 containing an electrode active material is applied in a constant thickness to a surface of the metallic foil 1 on which these processing is performed. Then, it is passed through a slurry drying furnace 9, and a solvent in the slurry 8 is removed, and the slurry is dried, and is wound round a reel 2b. When this plate is used, a paint film of the electrode active material does not fall in a battery assembling process.



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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the cell which has a plate for cells by the manufacture method and method of the plate for cells.

[0002]

[Description of the Prior Art] It had applied without pretreating conventionally the slurry which contained the electrode active material in the charge collector for the object for positive electrodes, or negative electrodes like a publication at JP,7-135023,A or JP,7-105937,A at all about manufacture of a rechargeable lithium-ion battery.

[0003]

[Problem(s) to be Solved by the Invention] However, in the plate produced by the manufacture method of the conventional plate for cells described above, the adhesion of a charge collector and an active material was bad, and exfoliated in the interface of a charge collector and an electrode active material during elevated-temperature preservation and repeat charge-and-discharge use, defluxion of an active material arose, and there was a problem of getting worse a fall and load characteristic of service capacity.

[0004]

[Means for Solving the Problem] this invention solves the above conventional technical problems, before it applies to a charge collector the slurry which contains an electrode active material as the manufacture method of the plate for cells, sprays a titania and the mixture of silicon system inorganic material on the surface of a charge collector, and, subsequently performs optical irradiation processing. According to this invention, an electrode active material exfoliates from a charge collector, it does not drop out, therefore there is neither a fall of the service capacity of a cell nor aggravation of a load characteristic.

[0005]

[Embodiments of the Invention] In the manufacturing method of the plate dried and manufactured after applying the slurry containing an electrode active material to the charge collector according to claim 1 which consists of a metallic foil like according to the manufacture method of the plate for cells of this invention By passing the process which precedes with the application of the aforementioned slurry and sprays a titania and the mixture of silicon system inorganic material on the surface of a charge collector, the process to dry, and the process which subsequently performs optical irradiation processing, and manufacturing a plate The cell which used this plate for cells can suppress small degradation of charge-and-discharge capacity, and degradation of a load characteristic in use of repeat charge and discharge.

[0006]

[Example] Hereafter, the example of this invention is explained. Drawing 1 is the model view of the manufacturing process of a plate. In drawing, 1 is a metallic foil for using as a charge collector of a positive electrode or a negative electrode, and this metallic foil 1 is equipment for spraying on the front face of a metallic foil 1 to which the speed of supply was supplied by part for 13m/from reel 2a, and 3 showed the atomiser to, and a titania and the mixture 4 of silicon system inorganic material were supplied from reel 2a. 5 is a dryer which dries the mixture 4 of the sprayed inorganic material, and although 6 shows an optical irradiation equipment and processed with corona discharge equipment in this example, sunlight and an effect with the same said of fluorescent lamp processing are acquired. Moreover, 7 is a coating machine, sprays a titania and the mixture 4 of silicon system inorganic material from an atomiser 3, and applies to fixed thickness the slurry 8 which contained the electrode active material after dryness on the front face of the metallic foil 1 by which corona discharge processing was carried out. When the metallic foil 1 to which this slurry 8 was applied passes the drying furnace 9 for slurries, the solvent in a slurry 8 is removed, a slurry dries, it is rolled round by reel 2b, and a plate is completed. The slurry 8 used by this example consists of an electrode active material, a binder, and a solvent, and solvents are a thickener and water. It mixed, respectively in the solution 45 weight section which dissolved in the styrene-butadiene-rubber 5 weight section as the carbon 50 weight section and a binder, and specifically dissolved the carboxyl methyl-cellulose 1 weight section in the water 99 weight section as a solvent as an electrode active material, and considered as the slurry. The obtained slurry is applied to a metallic foil using a coating machine 7.

[0007] Next, in this invention, a titania and the mixture 4 of silicon system inorganic material were sprayed, and by carrying out optical irradiation examined how the adhesive property of an electrode active material and a metallic foil 1 would be improved. The hydroxyl group OH has arrived at the front face, as shown in drawing 2 (a), if optical irradiation is performed here, an electron and a hole will generate the titania 10 which was sprayed on the front face of a metallic foil 1, and was dried,

and the mixture of the silicon system inorganic material 11 on a titania front face, an electron returns the oxygen in air, and a hole oxidizes and changes water into a hydroxyl group OH at super oxide ion (O<sub>2</sub><sup>-</sup>). Super oxide ion and a hydroxyl group OH are named generically, are called active oxygen, and have the very strong oxidative degradation force. This active oxygen decomposes the hydrophobic molecule R in the front face of a titania 10 and the silicon system inorganic material 11 into a carbon dioxide and water, as shown in drawing 2 (b), and it exposes a hydroxyl group OH. As shown in drawing 2 (c), the water in air sticks to the exposed hydroxyl group OH in an instant, and it is thought by water permeating the interior of the silicon system inorganic material 11 by surface diffusion that an adhesive property improves. Such surface change is observed in the form of increase of surface tension, and reduction of the degree of wetting angle. The result of the experiment was shown in Table 1. The degree of wetting angle (contact angle of water) used pure it. The method of Zismann estimated solid surface tension as a critical surface tension.

[0008]

[Table 1]

	水の接触角	臨界面張力 $\gamma^c$ (dyne/cm)
銅箔 (未処理)	49°	24
銅箔 (本発明)	7°	76
アルミニウム箔 (未処理)	54°	24
アルミニウム箔 (本発明)	21°	50

[0009] As shown in Table 1, the result with same copper foil and aluminum foil was obtained. That is, when both metallic foils carry out processing by this invention, compared with the case of being unsettled, the contact angle of water becomes small, and the critical surface tension is large. From this result, a titania and the mixture of silicon system inorganic material are sprayed, and by carrying out optical irradiation shows that the adhesion capacity of a surface of metal has been improved. [0010] Next, the cell was produced using the negative-electrode plate which sprayed the titania mentioned above and the mixture of silicon system inorganic material, and was produced using the metallic foil which carried out optical irradiation, and the cycle property was checked. Drawing 3 is the cross section of the cylindrical cell used for the example of this invention. The cell produced the thing of the diameter of 17mm, and height 50mm size. In drawing 3, the positive-electrode board with which 13 has the positive-electrode lead 14, the negative-electrode board with which 15 has the negative-electrode lead 16, the separator with which 17 intervenes between the positive-electrode board 13 and the negative-electrode board 15, and 18 are cell cases, and an up electric insulating plate and 19 obturate [ 18 ] a lower electric insulating plate and 20 with the obturation board 22 through a gasket 21. moreover -- as the example of comparison -- a mixture -- the cycle-life property of the cell using the plate which applied the paste to the charge collector of non-processed copper, and produced it, and this invention article is shown in drawing 4 When charge was performed by the 500mA constant current and set to 4.1V, it was switched to the constant-potential charge of 4.1V, and it performed charge for a total of 2 hours. When electric discharge was performed by 20 degrees C and 720mA and the discharge potential was set to 3.0V, electric discharge was ended and the next charge was started. This drawing 4 shows that degradation of capacity is excellent in a cycle property few, even if the cell of this invention repeats charge and discharge as compared with the cell of the conventional example. As for this, it is the cause that the mixture of a negative electrode sticks more with a charge collector, and stopped being able to separate easily also due to expansion contraction of the mixture in charge and discharge.

[0011] Moreover, after saving these cells for 60 degrees C and 20 days in the state of charge and performing charge and discharge several times in ordinary temperature after that, capacity until it discharges by 720mA and voltage amounts to 3.0V was calculated, and the rate over the capacity before the preservation was shown in Table 2.

[0012]

[Table 2]

	本発明	従来例
容量劣化率 = (保存後の放電容量) / (保存前の放電容量) × 100	96	90

[0013] Also in elevated-temperature preservation, this invention article became clear [ that degradation of capacity decreases ] as shown in Table 2.

[0014]

[Effect of the Invention] As explained above, according to this invention, in use of repeat charge and discharge, degradation of charge-and-discharge capacity and degradation of a load characteristic can be suppressed very small. Moreover, degradation of charge-and-discharge capacity and degradation of a load characteristic can be small suppressed under a severe condition which is left under an elevated temperature over a long period of time. Moreover, fall of the paint film of an inner electrode active material is lost as a cell erector, and he can improve workability.

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[Translation done.]

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CLAIMS

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[Claim(s)]

[Claim 1] The manufacture method of the plate for cells characterized by to have the process which sprays a titania and the mixture of silicon system inorganic material on the front face of the aforementioned charge collector in the manufacture method of the plate for cells dried and manufactured before the application process which applies the aforementioned slurry after applying the slurry containing an electrode active material to the charge collector which consists of a metallic foil, and subsequently performs optical irradiation processing.

[Claim 2] The cell which has the plate for cells characterized by having applied the slurry and being manufactured after carrying out optical irradiation of the titania prepared in the front face of the charge collector which consists of a metallic foil, and the mixture of silicon system inorganic material.

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[Translation done.]

**WEST**

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L5: Entry 7 of 31

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TITLE: MANUFACTURE OF ELECTRODE FOR BATTERY AND BATTERY HAVING ELECTRODE MADE BY THIS METHOD

PUBN-DATE: September 25, 1998

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APPL-NO: JP09051307

APPL-DATE: March 6, 1997

INT-CL (IPC): H01 M 4/66; H01 M 4/04; H01 M 10/40

## ABSTRACT:

PROBLEM TO BE SOLVED: To sharply restrain the degradation of charging-discharging capacity of a battery using this electrode and the degradation of a load characteristic by spraying titania and a silicon inorganic material on the surface of a collector, and performing light irradiating processing after it is dried before slurry containing an electrode active material is applied to the collector.

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## WEST



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L5: Entry 8 of 31

File: JPAB

Sep 22, 1998

PUB-NO: JP410249561A

DOCUMENT-IDENTIFIER: JP 10249561 A

TITLE: WELDING METHOD BETWEEN SAME KIND OF METAL, AND LI BATTERY HAVING POSITIVE ELECTRODE SEALING PLATE WITH POSITIVE REED WELDED BY SUCH METHOD

PUBN-DATE: September 22, 1998

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APPL-NO: JP09051308

APPL-DATE: March 6, 1997

INT-CL (IPC): B23 K 26/00; C23 G 5/00; H01 M 2/30

## ABSTRACT:

PROBLEM TO BE SOLVED: To enable the same kinds of metal to be welded to each other simply and firmly.

SOLUTION: A positive electrode sealing member A is fixed on its supporting body 10, an electrode 11 for corona discharge is fixed on the positive electrode sealing plate made of aluminum alloy plate to be processed, a corona discharge processing is done in a few seconds while a required voltage is applied, a face to be welded of the aluminum alloy plate of the positive electrode sealing plate 1 is processed to be a face free of impurities, and then, a positive electrode reed is firmly welded to the inner face of the positive electrode sealing plate 1 by laser welding.

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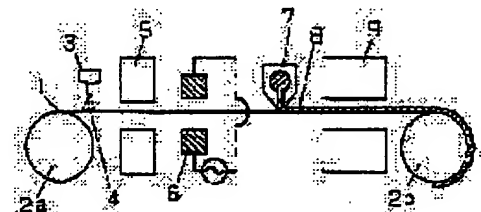
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OO FUMIO(54) MANUFACTURE OF ELECTRODE FOR BATTERY AND BATTERY HAVING ELECTRODE  
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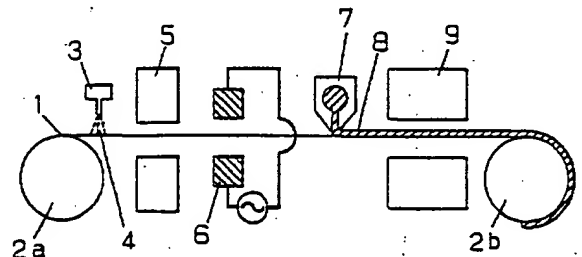
(54) 【発明の名称】 電池用極板の製造方法とその方法による電池用極板を有する電池

(57) 【要約】

【課題】 繰り返し充放電の使用において、充放電容量の劣化や負荷特性の劣化を極めて小さく抑制した電池を提供することを課題とする。

【解決手段】 極板の製造工程として金属箔1よりなる集電体に電極活性物質のスラリー8を塗布するのに先行して、チタニアおよびシリコン系無機材料の混合物4を噴霧する工程と、乾燥処理を行う工程と、次いで光照射装置6によって光照射する工程とを有する極板の製造方法および上記製造方法による極板を提供し、充放電容量の劣化や負荷特性の劣化の小さい電池を実現する。

- 1 金属箔
- 2 a, 2 b リール
- 3 噴霧装置
- 4 混合物
- 5 乾燥装置
- 6 光照射装置  
(コロナ放電装置)
- 7 コーター
- 8 スラリー
- 9 スラリー用乾燥炉



## 【特許請求の範囲】

【請求項1】 金属箔よりなる集電体に電極活物質を含有するスラリーを塗布した後に乾燥して製造する電池用極板の製造方法において、前記スラリーを塗布する塗布工程前に前記集電体の表面にチタニアとシリコン系無機材料の混合物を噴霧し、次いで光照射処理を行う工程を有することを特徴とする電池用極板の製造方法。

【請求項2】 金属箔よりなる集電体の表面に設けたチタニアとシリコン系無機材料の混合物を光照射した後、スラリーを塗着して製造されたことを特徴とする電池用極板を有する電池。

## 【発明の詳細な説明】

## 【0001】

【発明の属する技術分野】本発明は、電池用極板の製造方法とその方法による電池用極板を有する電池に関する。

## 【0002】

【従来の技術】従来、リチウムイオン二次電池の製造に関しては、正極用または負極用の集電体に、特開平7-135023号公報または特開平7-105937号公報に記載のように、電極活物質を含んだスラリーを何らかの前処理せずに塗布していたものであった。

## 【0003】

【発明が解決しようとする課題】しかしながら、前記する従来の電池用極板の製造方法により作製した極板においては、集電体と活物質の密着性が悪く、高温保存中や、繰り返し充放電使用中に集電体と電極活物質との界面で剥離し、活物質の脱落が生じ、放電容量の低下や負荷特性を悪化するという問題があった。

## 【0004】

【課題を解決するための手段】本発明は上記のような従来の課題を解決するもので、電池用極板の製造方法として電極活物質を含有するスラリーを集電体に塗布する前に、集電体の表面にチタニアとシリコン系無機材料の混合物を噴霧し、次いで光照射処理を施したものである。本発明によれば、電極活物質が集電体より剥離したり脱落したりすることがなく、従って電池の放電容量の低下や負荷特性の悪化がない。

## 【0005】

【発明の実施の形態】本発明の電池用極板の製造方法によれば、請求項1記載のように金属箔よりなる集電体に電極活物質を含有するスラリーを塗布した後に乾燥して製造する極板の製造法において、前記スラリーの塗布に先行して集電体の表面にチタニアとシリコン系無機材料の混合物を噴霧する工程と、乾燥する工程と、次いで光照射処理を行う工程とを経過して極板を製造することにより、この電池用極板を使用した電池は繰り返し充放電の使用において、充放電容量の劣化や負荷特性の劣化を小さく抑制することができる。

## 【0006】

【実施例】以下、本発明の実施例を説明する。図1は、極板の製造工程のモデル図である。図において、1は正極または負極の集電体として用いるための金属箔であり、この金属箔1はリール2aから供給速度が13m/分で供給され、また3は噴霧装置を示し、チタニアとシリコン系無機材料の混合物4を、リール2aから供給された金属箔1の表面に噴霧するための装置である。5は噴霧された無機材料の混合物4を乾燥させる乾燥装置であり、6は光照射装置を示すもので本実施例ではコロナ放電装置により処理を行ったが、太陽光や蛍光灯処理でも同様な効果が得られる。また7はコーターであり、チタニアとシリコン系無機材料の混合物4を噴霧装置3より噴霧し、乾燥後、コロナ放電処理された金属箔1の表面に電極活物質を含んだスラリー8を一定の厚みに塗布するものである。このスラリー8を塗布された金属箔1は、スラリー用乾燥炉9を通過することにより、スラリー8中の溶媒が除去され、スラリーが乾燥し、リール2bに巻き取られ極板が完成される。本実施例で用いたスラリー8は、電極活物質、バインダーおよび溶媒からなり、溶媒は増粘剤と水である。具体的には電極活物質としてカーボン50重量部、バインダーとしてスチレンブタジエンゴム5重量部、溶媒としてカルボキシルメチルセルロース1重量部を水99重量部に溶解した水溶液45重量部にそれぞれ混合しスラリーとした。得られたスラリーをコーター7を用いて金属箔に塗布する。

【0007】次に、本発明においてチタニアとシリコン系無機材料の混合物4を噴霧し、光照射することにより、電極活物質と金属箔1の接着性がいかに改善されるかを検討した。図2(a)に示すように金属箔1の表面に噴霧、乾燥されたチタニア10とシリコン系無機材料11の混合物は、表面に水酸基OHが着いており、ここに光照射を行うとチタニア表面に電子とホールが発生し、電子は空気中の酸素を還元してスーパーオキシドイオン( $O_2^-$ )に、ホールは水を酸化して水酸基OHに変える。スーパーオキシドイオンと水酸基OHは総称して活性酸素と呼ばれ、非常に強い酸化分解力を持つ。この活性酸素が、図2(b)に示すようにチタニア10とシリコン系無機材料11の表面にある疎水性分子Rを二酸化炭素と水に分解し、水酸基OHをむき出しにさせる。図2(c)に示すように、露出した水酸基OHには空気中の水が瞬時に吸着し、表面拡散によりシリコン系無機材料11の内部に水が浸透することで、接着性が改善するものと考えられる。このような表面変化は、表面張力の増大や、濡れの角度の減少という形で観測される。表1にその実験の結果を示した。濡れの角度(水の接触角)は純粋のそれを用いた。固体の表面張力は、Zismanの方法によって臨界表面張力として評価した。

## 【0008】

## 【表1】

	水の接触角	臨界面張力 $\gamma^c$ (dyne/cm)
銅箔（未処理）	49°	24
銅箔（本発明）	7°	76
アルミニウム箔（未処理）	54°	24
アルミニウム箔（本発明）	21°	50

【0009】表1からわかるように、銅箔およびアルミニウム箔とも同様な結果が得られた。すなわち、両金属箔とも本発明による処理をした場合は、未処理の場合に比べて水の接触角は小さくなり、臨界面張力は大きくなっている。この結果からチタニアとシリコン系無機材料の混合物を噴霧し、光照射することにより、金属表面の接着能力が改善されたことがわかる。

【0010】次に、上述したチタニアとシリコン系無機材料の混合物を噴霧し、光照射した金属箔を用いて作製した負極極板を用いて電池を作製し、そのサイクル特性を確認した。図3は本発明の実施例に用いた円筒型電池の断面図である。電池は、直径17mm、高さ50mmのサイズのものを作製した。図3において、13は正極リード14を有する正極板、15は負極リード16を有する負極板、17は正極板13と負極板15間に介在するセパレータ、18は上部絶縁板、19は下部絶縁板、20は電池ケースで、ガスケット21を介して封口板22により封口したものである。また比較例として、合剤

ペーストを無処理の銅の集電体に塗布して作製した極板を用いた電池と本発明品のサイクル寿命特性を図4に示す。充電は500mAの定電流で行い4.1Vになった時点で4.1Vの定電圧充電に切り換え、合計2時間充電を行った。放電は、20℃、720mAで行い、放電電位が3.0Vになった時点で放電を終了し次の充電を開始した。この図4より本発明の電池は従来例の電池と比較して、充放電を繰り返しても容量の劣化が少なくサイクル特性に優れていることがわかる。これは負極の合剤が集電体と、より密着し充放電での合剤の膨張収縮によっても剥れにくくなったことが原因である。

【0011】また、これらの電池を充電状態で60℃、20日間保存し、その後、常温にて数回充放電を行った後、720mAで放電を行い電圧が3.0Vに達するまでの容量を求め、その保存前の容量に対する割合を表2に示した。

【0012】

【表2】

	本発明	従来例
容量劣化率＝ (保存後の放電容量) / (保存前の放電容量) ×100	96	90

【0013】表2に示す通り、高温保存においても本発明品は容量の劣化が少なくなることが明らかとなった。

【0014】

【発明の効果】以上説明したように、本発明によれば、繰り返し充放電の使用において、充放電容量の劣化や負荷特性の劣化を極めて小さく抑制することができる。また、高温下に長期放置するような厳しい条件下においても、充放電容量の劣化や負荷特性の劣化を小さく抑制することができる。また、電池組立工程中における電極活物質の塗膜の落下がなくなり、作業性を改善することが

できる。

【図面の簡単な説明】

【図1】本発明の一実施例における電池用極板の製造工程を示す略図

【図2】金属箔の表面の改質状態を示すモデル図

【図3】本発明に用いた電池の断面図

【図4】サイクル寿命特性を比較した図

【符号の説明】

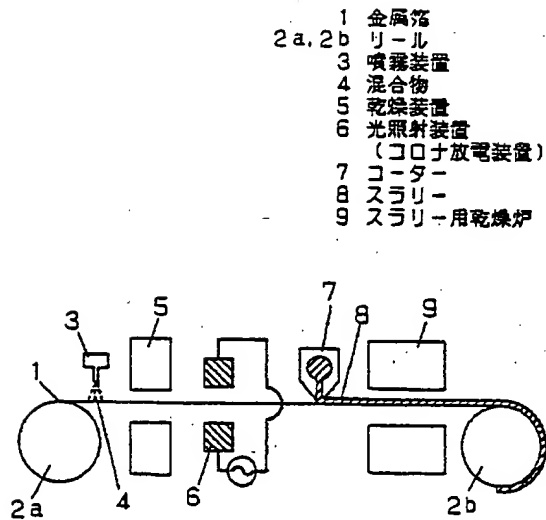
1 金属箔

2 a, 2 b リール

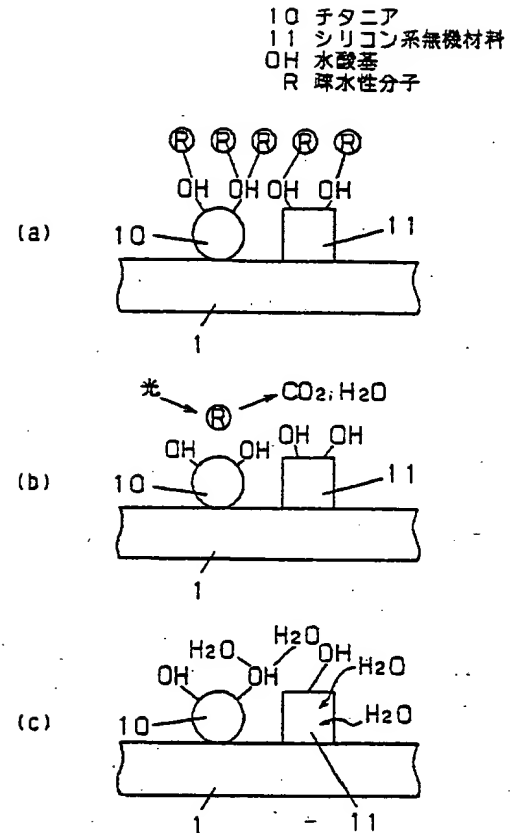
- 3 噴霧装置
- 4 混合物
- 5 乾燥装置
- 6 光照射装置 (コロナ放電装置)
- 7 コーター
- 8 スラリー

- 9 スラリー用乾燥炉
- 13 正極板
- 15 負極板
- 17 セパレータ
- 20 電池ケース
- 22 封口板

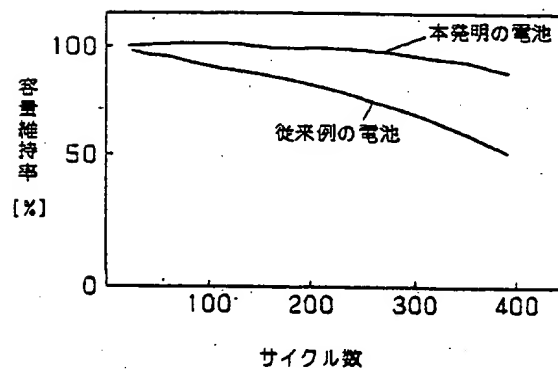
【図1】



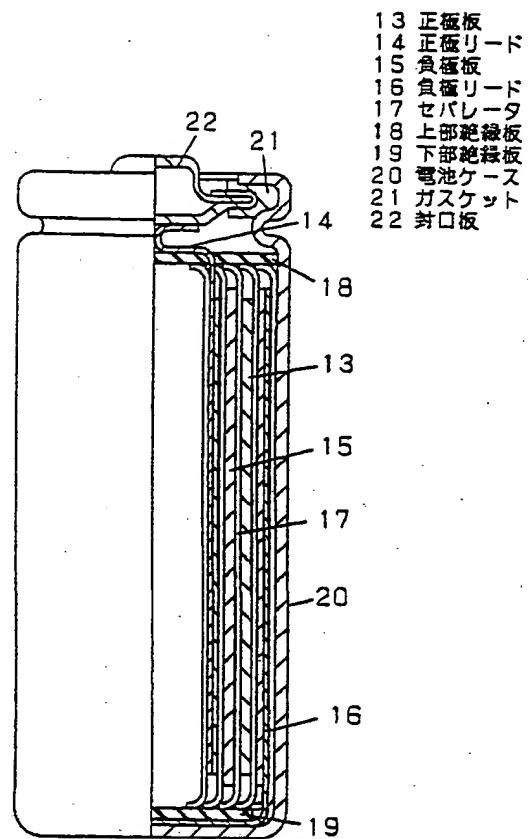
【図2】



【図4】



【図3】



フロントページの続き

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